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In this project we have performed both experimental and theoretical studies of optical beam phase-conjugation and of electromagnetic scattering and propagation with intense optical fields. During the reporting period we have: (1) demonstrated that atomic vapors require fewer photons to perform optical wave mixing than any other medium examined to date; (2) showed theoretically that a one-joule broadband optical pulse, whose carrier wavelength is one micron, can impart nearly one GeV energy to a charged particle; (3) established the stringent experimental upper limits on the hyperpolarizabilities of C<sub>60</sub> and C<sub>70</sub> molecules in solution; (4) made the first direct time-of-flight measurements of the drift velocity of photoexcited carrier in any photorefractive insulator (n-type Bi<sub>12</sub>SiO<sub>20</sub>). (5) determined the difference between the complex polarizabilities of different trap levels in insulators; (6) made quantitative predictions and measurements of spatial harmonic content of photorefractive gratings; (7) developed and applied moving-grating diagnostic techniques to photoexcited carriers; (8) demonstrated an exception to the law of exponential attenuation of weak monochromatic optical beams in a homogeneous medium.

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## 1. INTRODUCTION AND PROJECT OBJECTIVES

Nonlinear optics, the study of matter interactions with intense electromagnetic fields, has uncovered surprising and useful physical effects, such as stimulated scattering of light by which an intense monochromatic beam can be converted to intense coherent beams of different wavelengths. Recently, interesting processes involving nonlinear optical image-bearing beams have been discovered, such as optical-beam phase conjugation. Optical-beam phase conjugation is the name given to any process which generates, in real time, the time-reversed replica of a complex, image-bearing, optical beam, or which generates other related reflected beams, both monochromatic and polychromatic. Phase conjugation by optical four-wave mixing was conceived earlier in this project, as were such applications of phase conjugation as the corrections of image aberrations, brightness enhancement of laser outputs, edge-enhancements of images, automatic steering of beams, and optical spectroscopy by Raman-induced phase conjugation (RIPC). Such nonlinear optical techniques are currently being extended to perform many forms of beam processing, sorting, and routing on picosecond time scales. This project aims at exploring and developing these and other new wave-mixing processes, and the necessary nonlinear materials. Other novel electromagnetic scattering processes which occur at intense optical fields available from lasers are also explored for their scientific and device interest. The approaches taken in this project are both experimental and theoretical.

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## **2. MAJOR ACCOMPLISHMENTS: 12/1/90 -11/30/93**

The major accomplishments of this grant period fall into five areas of nonlinear optics: 1) the use of atomic vapors as highly nonlinear media for performing optical image processing and logic: "Atoms for logic"; 2) the discovery of optical pulse solutions of Maxwell's equations in vacuum which have an extraordinary ability to accelerate charge; 3) the measurement of the nonlinear and linear refractive indices in films and solutions of novel optical polymers and fullerene molecules; 4) the characterization and physics of photoconductivity in photorefractive single crystals; and 5) the demonstration and analysis of the anomalous propagation of light in highly scattering random media. Highlights of these accomplishments were 1) the demonstration of atomic vapor media that require fewer photons to perform optical mixing than any other medium ever examined; 2) the demonstration that a one-joule broadband optical pulse around one-micron wavelength can impart  $\sim$  one GeV energy to a charge; 3) the establishment of the best experimental upper limit on the hyperpolarizability of  $C_{60}$  and  $C_{70}$  molecules in solution; 4) the first direct time-of-flight measurement of the drift velocity of photo-excited carriers in any photorefractive insulator (n-type cubic  $Bi_{12}SiO_{20}$ ); 5) the first determination of the difference between the complex optical polarizabilities of occupied and unoccupied deep traps in a photorefractive insulator; 6) the first quantitative measurements of the spatial harmonic content of photorefractive charge and index gratings; 7) the development of a simple all-optical technique for measuring the real and imaginary parts of both spatial components (in-phase and in-quadrature) of any optical dielectric tensor grating induced by an optical intensity grating; and 8) the first observation of the non-exponential attenuation of a weak optical beam in a homogenous medium, the first exception to the attenuation law first enunciated by Pierre Bouguer 260 years ago. We outline these and the other accomplishments along with the pertinent published references in sections 2.1 to 2.5 below.

### **2.1 Atoms for logic**

In the 1990 proposal for this project, we argued that cesium or other alkali atoms in a vapor would exhibit larger optical nonlinearity than other materials had shown to date, and would exhibit this nonlinearity at diode-laser wavelengths and powers. We have verified this hypothesis and begun construction of a  $10^7$  pixel

image correlator employing a thin alkali vapor cell. The correlator is essentially the same as that designed thirty years ago by vander Lugt to recognize objects hidden in a noisy scene (Van64). However vander Lugt's static hologram is replaced by the fast-responding atoms that can form a new hologram every 100 ns. This arrangement is illustrated in Figs. 2.1.1a and 2.1.1b. Our correlator is essentially the same as that demonstrated by White and Yariv (Whi80) who used a photorefractive crystal as the nonlinear element. However our version, with an atomic-vapor as the nonlinear medium, can process two orders-of-magnitude more pixels because the medium is thinner than the distance for a pixel to diffract. Also our version correlates at seven orders-of-magnitude greater frame rate, and requires four orders-of-magnitude fewer photons per pixel to correlate.

We constructed and evaluated K and Cs vapor cells for their ability to perform in a correlator by measuring degenerate four-wave mixing (DFWM) in these cells as function of the input beam angle (at the resonant wavelengths of 770 and 852 nm respectively). Our results for 50 micron thick cells are shown in Fig. 2.1.2 along with the best competing results (Wan92, Par93) which were obtained with multi-quantum well layers (the closest solid state analogue to an atomic vapor). Conditions under which these results were obtained are given in Table 2.1.1 along with a comparison figure-of-merit derived as follows.

The prime "figure-of-merit" for comparing nonlinear media for correlator applications is the number of photons per pixel  $P_{pp}$  required to achieve nearly 100 % diffraction of beam H in Fig. 2.1.1. (The fewer photons per pixel  $P_{pp}$ , the better.) This  $P_{pp}$  turns out to be related to the DFWM data essentially as follows.

In DFWM two beams of intensity  $\frac{1}{2}I$  are incident at angles  $\pm\theta$  to the medium face, and create an index grating of spatial period  $\Lambda$  after time  $\tau$  in the medium.

$\Lambda = \frac{1}{2}\lambda / \sin \theta$ . A phase-matched beam scatters from this grating with efficiency  $R$ , at small  $\theta$  that is proportional to  $I^2$  in the operating regime. At some beam half angle  $\theta_a$  the efficiency  $R$  falls to half its small angle value. The number of pixels that can be correlated is approximated well by the value of  $[\text{beam area} / \Lambda_a^2]$ , where  $\Lambda_a \equiv \Lambda(\theta = \theta_a)$ . The energy  $U$  required is  $[I \times \text{beam area} \times \tau]$  and the number of photons is  $U\lambda / hc$ . Therefore the number of photons per pixel to achieved efficiency  $R \sim 1$  is

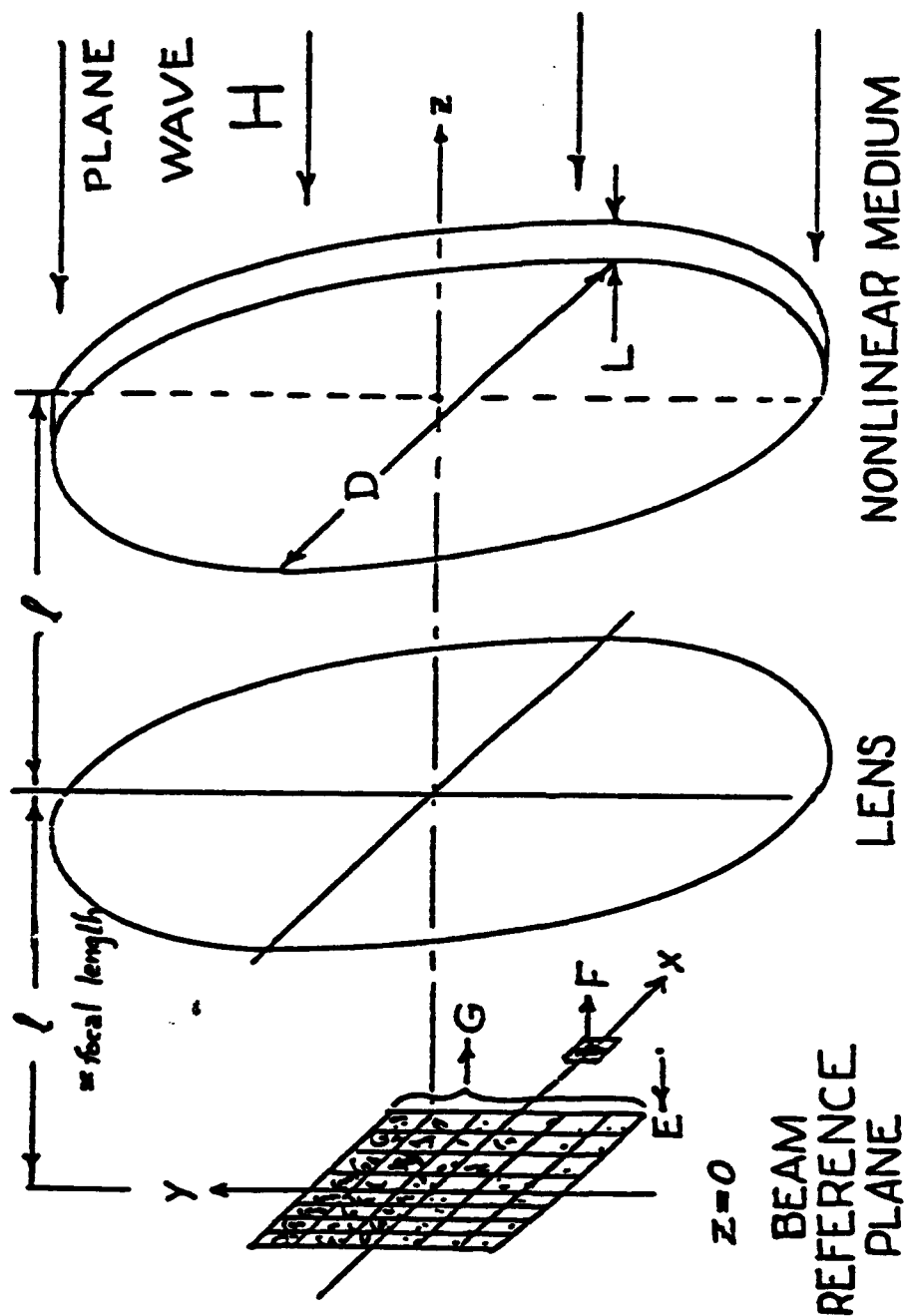


Fig. 2.1.1a. Schematic of the correlator showing: (1) right-traveling beam  $G$  bearing array of items in memory, (2) right-traveling beam  $F$  bearing item to be compared to memory items, and (3) left-traveling output beam  $E$  bearing correlation signal (if one exists) and background.

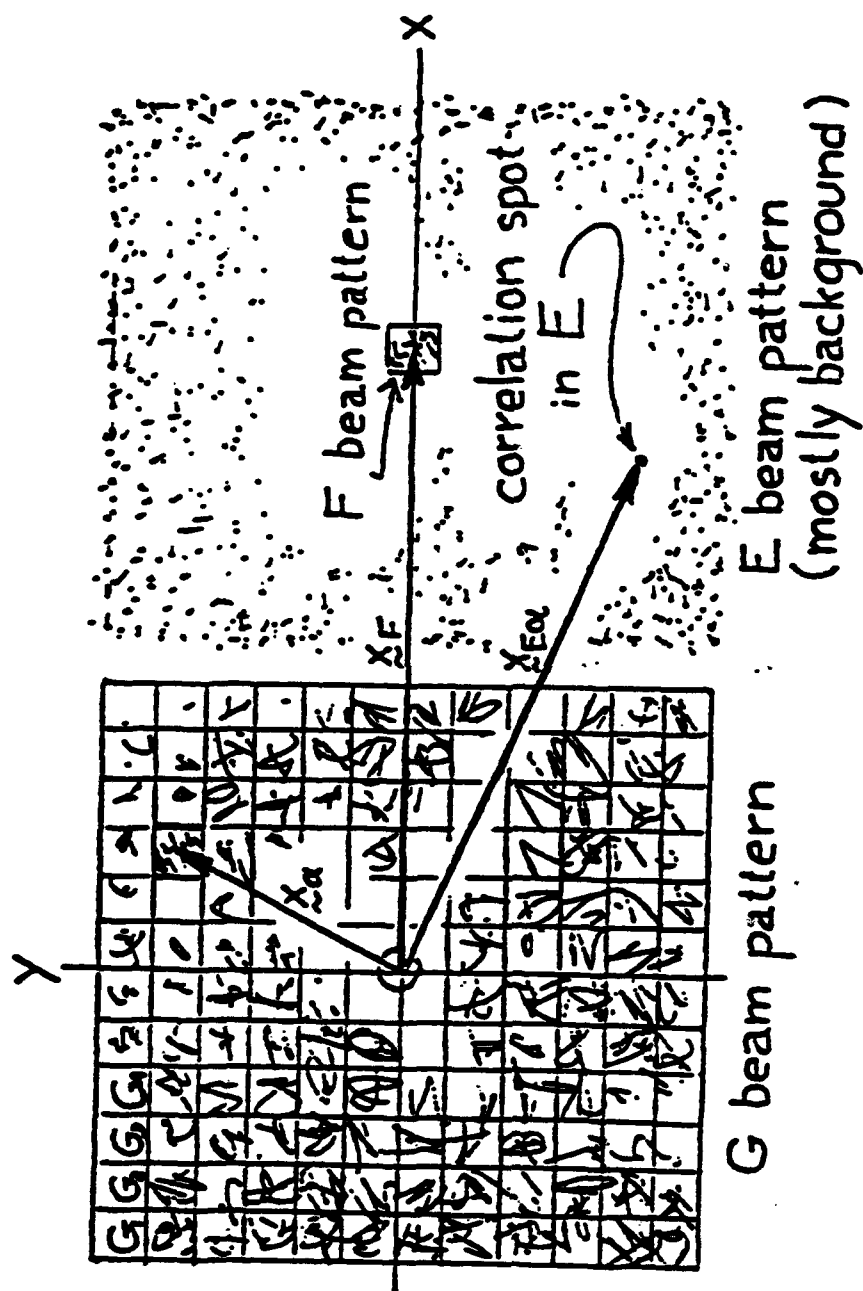


Fig. 2.1.1b. Schematic of beam field patterns in the reference plane ( $z=0$ ).

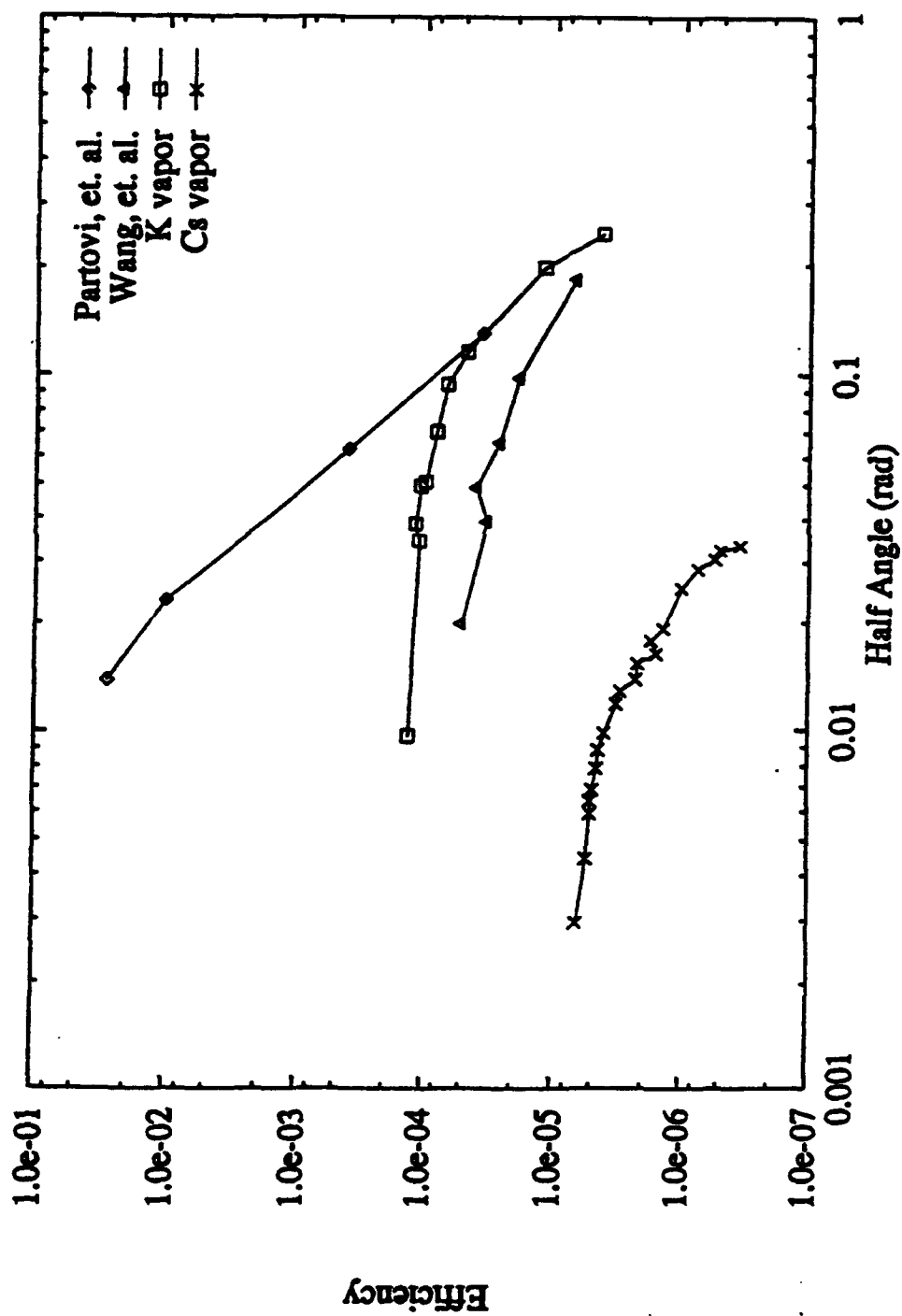


Fig. 2.1.2. Diffraction efficiency versus angle in experiments with photorefractive quantum wells and atomic vapors as nonlinear media.

$$P_{pp} = I\tau\lambda^3 / (4R_0^{\frac{1}{2}}hc \sin^2 \theta_a), \quad (2.1.1)$$

where  $R_0$  is the actual diffraction efficiency for small beam angle  $\theta (<< \theta_a)$  at the experimental intensity  $I$ . This relation gives the photons-per-pixel values  $P_{pp}$  of Table 2.1.1. Unfortunately, definite values for response time  $\tau$  and experimental intensity  $I$  were not given in (Wan92), but they are probably close to those in the experiments of (Par93). Therefore we have good reason to believe that the atomic vapor medium not only can correlate faster than multi-quantum-well structures but can do so with fewer photons per pixel.

Table 2.1.1. The number of photons  $P_{pp}$  required to correlate one pixel with near unity efficiency, and experimental parameters used in Eq. (2.1.1) for the estimate. The multi-quantum-well material MQW was described in (Par93). The K and Cs vapor data are from this project.

	$\lambda$ nm	$\theta_a$ mr	$I$ W/cm <sup>2</sup>	$\tau$ ns	$R_0$ %	$\Lambda_a$ $\mu$ m	$P_{pp}$
MQW	830	20	0.28	3000	2.8	21	$9 \times 10^7$
K	770	95	380	0.3	0.013	3.9	$6 \times 10^6$
Cs	852	11	0.01	30	0.0007	39	$7 \times 10^6$

The above results from this project are reported in the published summaries for Talks 4.28 and 4.29 below. However, we believe we can lower our photons-per-pixel number  $P_{pp}$  by at least an order of magnitude in the future. In addition, we propose work to speed the imminent realization of a spatial light modulator, needed to introduce the images into the correlator, whose speed and capacity will approach that of the vapor cell. The following results will aid vapor cell improvements.

**2.1.1 Optical pumping.** Optical pumping often limits the transitions, which can be used for near- or on- resonant degenerate four wave mixing in atomic vapors, to the "cycling" ones. We have shown experimentally that, by applying a second, state-preparation laser, noncycling transitions can be used for nonlinear response. This second laser restores the interaction between the three incoming beams from the main laser and the atoms by pumping the atoms back to the desired level. These results are reported in Pub. 3.12 and in Talk 4.14.

We have also demonstrated experimentally that frequent wall collisions in thin cells relax the optically pumped atoms back to their thermal distribution greatly strengthening the four-wave-mixing signal in some cases. Preliminary results of this research were reported in Talk 4.15 with a published summary.

## **2.2 Acceleration of charged particles by an electromagnetic pulse in vacuum**

A one Joule focused optical pulse, 90% of whose spectral content is at wavelengths longer than one micron, can impart nearly one GeV of energy to a relativistic particle of charge  $e$  moving with the pulse, along its axis, up to the focal point in vacuum. The simple solutions of Maxwell's equations which have this property were discovered by Ziolkowski (Zio89) who called them "MPS pulses". In this project we achieved the first correct calculation 1) of the energy contained in these solutions, 2) of their spectral content, and 3) showed that a certain subset of these solutions had an unexpectedly high ability to accelerate charge. We have named this special subset of these solutions "focused doughnut" pulses. See Talk 4.12 below; a long publication is in preparation. Our results for focused doughnut pulses are summarized here.

The electric and magnetic fields  $\vec{E}(\vec{x}, t)$  and  $\vec{H}(\vec{x}, t)$  of a transverse electric (TE) focused doughnut pulse can be written simply (in SI units) as

$$\vec{E} = -\mu_0 \text{curl}(\partial / \partial t) \hat{z} f(\vec{x}, t), \quad (2.2.1)$$

and

$$\vec{H} = \text{curl} \text{curl} \hat{z} f(\vec{x}, t),$$

where  $\mu_0$  is the permeability of vacuum and

$$f(\vec{x}, t) = f_0 [c^2 t^2 - r^2 - iz(q_2 - q_1) + ict(q_2 + q_1) - q_1 q_2]^{-1}. \quad (2.2.2)$$

Here  $f_0$ ,  $q_2$  and  $q_1$  are adjustable real constants and  $r^2 = x^2 + y^2 + z^2$ . We can, without loss of generality, take  $q_2$  and  $q_1$  to be positive and  $q_2 \geq q_1$ . The real and imaginary parts of (2.2.1) are linearly independent TE solutions, and the dual of these solutions are transverse magnetic (TM) pulse solutions. All four solutions have the focused doughnut energy distribution illustrated schematically in Fig. 2.2.1. We showed that the energy  $U$  of each of these four pulse solutions is

$$U = \frac{\pi^2 \mu_0 f_0^2}{4 q_1^2 q_2^3} \left(1 + \frac{q_2}{q_1}\right) \quad (2.2.3)$$

and is decomposable into an energy spectrum:

$$u(\omega) \propto \omega^4 a^{-3} (a \cosh a - \sinh a) \exp[-\omega(q_1 + q_2)/c], \quad (2.2.4)$$

where

$$a = (q_2 - q_1)\omega/c. \quad (2.2.5)$$

We also extended these results to the full 3-parameter ( $q_1$ ,  $q_2$  and  $q_3$ ) MPS solution (Zio89).

The acceleration of a charge by a focused doughnut pulse is greatest for the transverse magnetic (TM) imaginary solution  $E_{TM}''(\vec{x}, t)$  if one assumes (as we believe) that the acceleration is greatest for the geometry shown in Fig. 2.2.2. That is, the charge travels with velocity near the velocity of light  $c$  along the axis so as to arrive with the pulse at the center point of maximum focus where it is decoupled, as by the plasma/metal wall shown in Fig. 2.2.2. The energy  $\Delta W$  imparted to the charge is

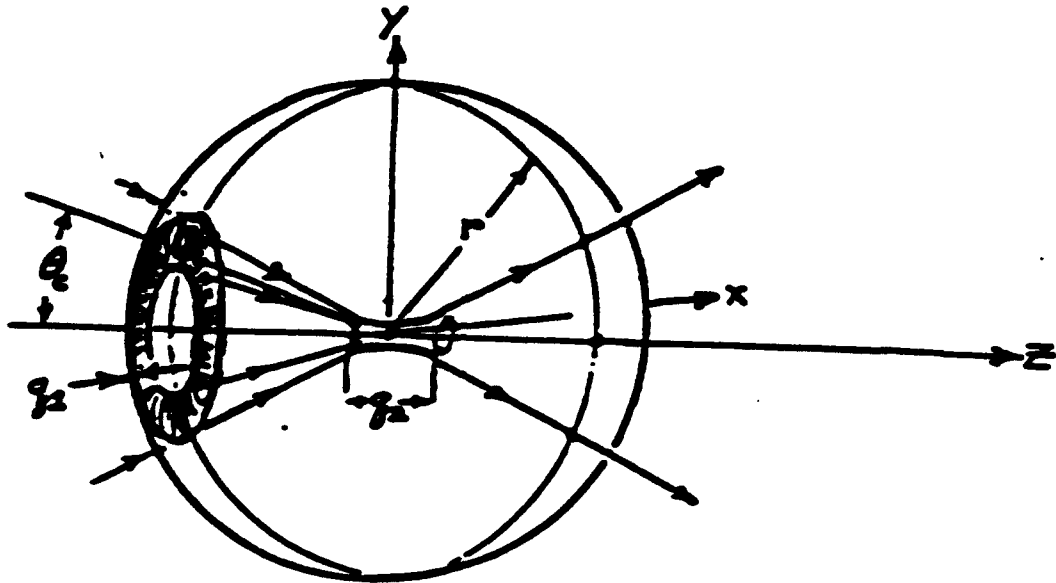


Fig. 2.2.1. Schematic of "focused doughnut" pulse at time  $t = -r/c$ . Maximum focusing occurs at  $t = 0$  at origin of coordinates. The pulse thickness  $q_1$  and Rayleigh length  $q_2$  of focal region are indicated. As for focused cw gaussian

beams, the divergence half angle  $\theta_c$  (i.e., the cone angle) equals  $\sqrt{q_1/q_2}$ .

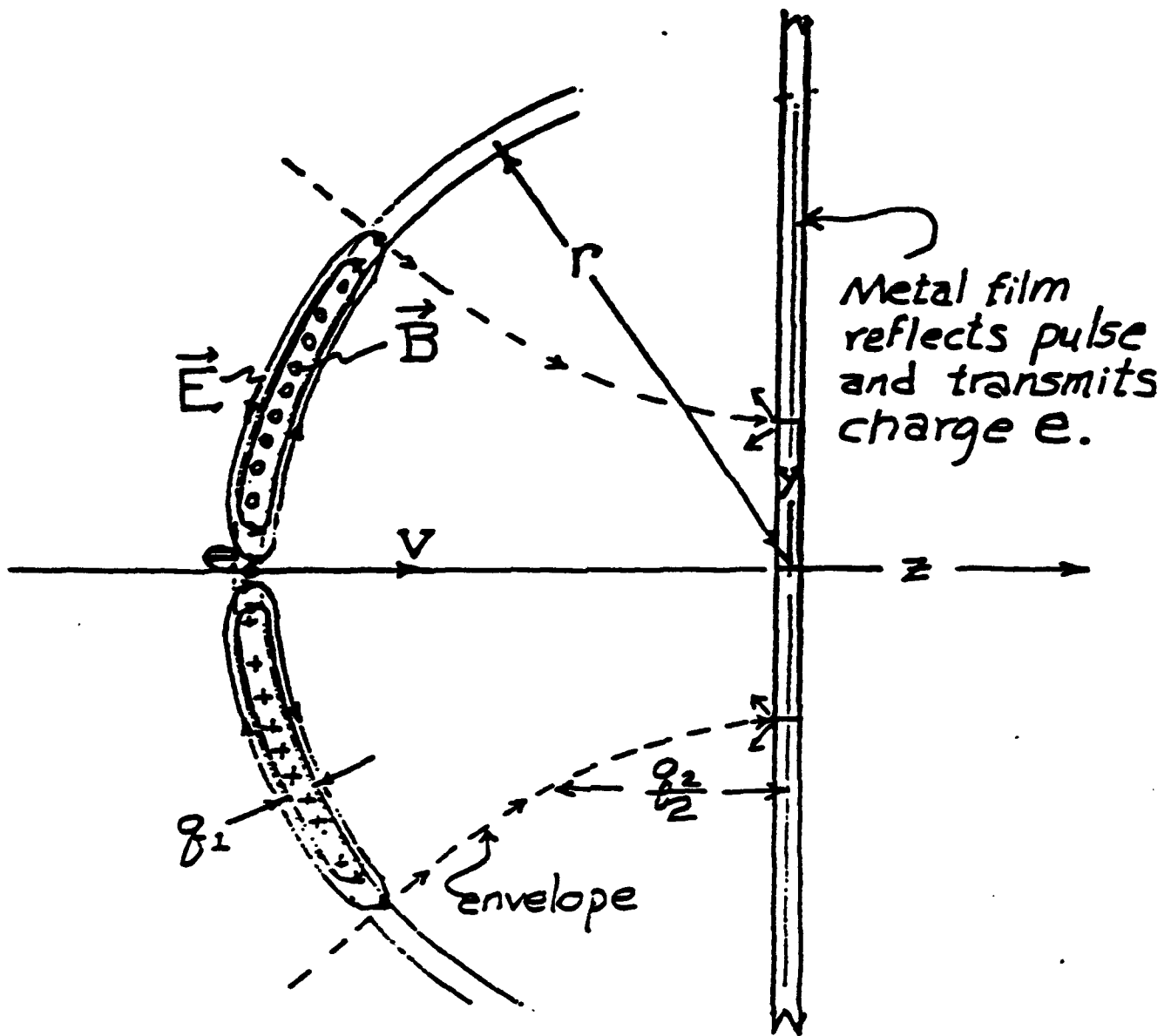


Fig. 2.2.2. Schematic of TM "focused doughnut" pulse accelerating charge  $e$ , having velocity  $v$ , as maximum focus is approached. The picture here is at time  $r/c$  before maximum focus at the origin of coordinates. The characteristic length parameters  $q_1$  and  $q_2$  which completely specify the solution are indicated.

$$\Delta W = ec \int_{-\infty}^0 dt E_{TM}''(0,0,ct,t) = 2ef_0(\mu_0/\epsilon_0)^{\frac{1}{2}} q_1^{-2} q_2^{-1}, \quad (2.2.6)$$

where  $\epsilon_0$  is the permittivity of vacuum. It is this result that is the basis for the "one Joule at one micron imparts one GeV" rule-of-thumb that we quoted above.

### 2.3 Optical studies of fullerenes and polymers

During this project period, we built apparatus to synthesize and purify  $C_{60}$  and other fullerenes. We performed many measurements of the linear  $\chi$  and nonlinear  $\chi^{(3)}$  optical response of these fullerenes, both in solutions and on solid films formed on glass. We used a simple model to calculate the low frequency limit of  $\chi$  and of  $\chi^{(3)}$ , and we found the allowed rotational states of the free fullerene  $^{12}C_{60}$  using group theory. We also measured the logarithmic decrement in transmission of a weak 532 nm pulse through  $C_{60}$ /benzene solutions at times up to 10 ns after passage of a 30 ps, 532 nm pulse having irradiance in the range 0.2 to 10 mJ/cm<sup>2</sup>. We conclude that the large decrease in transmission we observe is indeed mostly due to excitation of an excited (highly absorbing) long-lived triplet electronic state of  $C_{60}$ , just as Tutt and Kost (Tut92) hypothesized to explain the reverse saturable absorption they observed for single 10 ns pulses in similar solutions. However, the parameters we deduce predict somewhat different nanosecond characteristic cross-sections than are deduced from the nanosecond experiments in (Tut92) and also in (Jos93). In our view, this strong reverse saturable absorption is a very promising optical property for practical applications, as we will explain below and in Sec.3., so we propose to understand it in detail.

We also continued our collaboration with L. Dalton of the USC chemistry department on development of optical polymers.

**2.3.1 Hyperpolarizability of  $C_{60}$  and  $C_{70}$ .** We have performed four-wave-mixing experiments with  $C_{60}$ /benzene solutions and determined that the upper limit of the real part of the hyperpolarizability of the  $C_{60}$  molecule is 60 times larger than the one of the benzene molecule. This upper limit is barely consistent with the experimental value reported for a thin film sample in Ref. Kaf92, provided that the relative large error margins in both experiment are taken

into account. Our experiments and analysis are described in Pub. 3.20.

Unfortunately, we found conclusively that C<sub>60</sub> and C<sub>70</sub> have orders-of-magnitude lower hyperpolarizability ( $\chi^{(3)}$  at long wavelength) than was suggested in the early experiments of Blau, *et al.* (Bla91). Consequently, C<sub>60</sub> and C<sub>70</sub> are not promising as  $\chi^{(3)}$  nonlinear optical materials. See the summary of our Talk 4.17. In the course of these measurements, we did make the most accurate comparison to date between hyperpolarizabilities of carbon disulfide and benzene, yielding the most accurate value to date for the  $\chi^{(3)}$  of benzene at long wavelengths. See Pub. 3.20.

**2.3.2 Theoretical studies.** We completed the calculation of the linear polarizability  $\alpha$  and third-order nonlinear polarizability  $\gamma$  at zero frequency of 60 spin-paired, non-interacting electrons confined to a sphere of radius  $r$  and in their ground state. (Pub. 3.21.) This is a straightforward extension of the theory of Rustagi and Ducuing (Rus74) for  $\alpha$  and  $\gamma$  of  $n$  free electrons on a wire of given length. Curiously, our calculation predicts a negative  $\gamma$  for C<sub>60</sub> whereas all experiments to date and all exact calculations (done for H, He and Li) have produced positive  $\gamma$ . We suspect the negative sign is simply an indication that Coulomb interactions play an important role and will reverse the sign of  $\gamma$  when added.

We also calculated the allowed rotational states of the free <sup>12</sup>C<sub>60</sub> fullerene molecule. Since each <sup>12</sup>C atom is a boson, only those rotational states that are even under interchange of any carbon coordinates are allowed. We found that only states of angular momentum 0, 6, 10, ... are allowed. As high angular momentum is approached, only one in 60 of the states allowed without bose symmetry survives. These results were described in Talk 4.11 and are in manuscript.

**2.3.3 Reverse saturable absorption in C<sub>60</sub> solutions.** Excited by the report of Tutt and Kost (Tut92) of large reverse saturable absorption in C<sub>60</sub> solutions, we measured the optical transmission experienced by a weak 532 nm, 30 ps pulse in C<sub>60</sub>/benzene solutions at variable time  $t$  after a "strong" 532 nm, 30 ps saturating pulse. Typical results for strong-pulse fluences from 0.2 to 10 mJ/cm<sup>2</sup> are shown for  $0 < t < 200$  ps in Fig. 2.3.1. The reduced transmission observed there stays constant for delay times  $t$  up to the limit (10 ns) of our apparatus. This is consistent with the excitation of a strongly absorbing triplet state whose lifetime  $t_T$  is known to be longer than 10 ns, even in impure samples (Tut92). Consider the

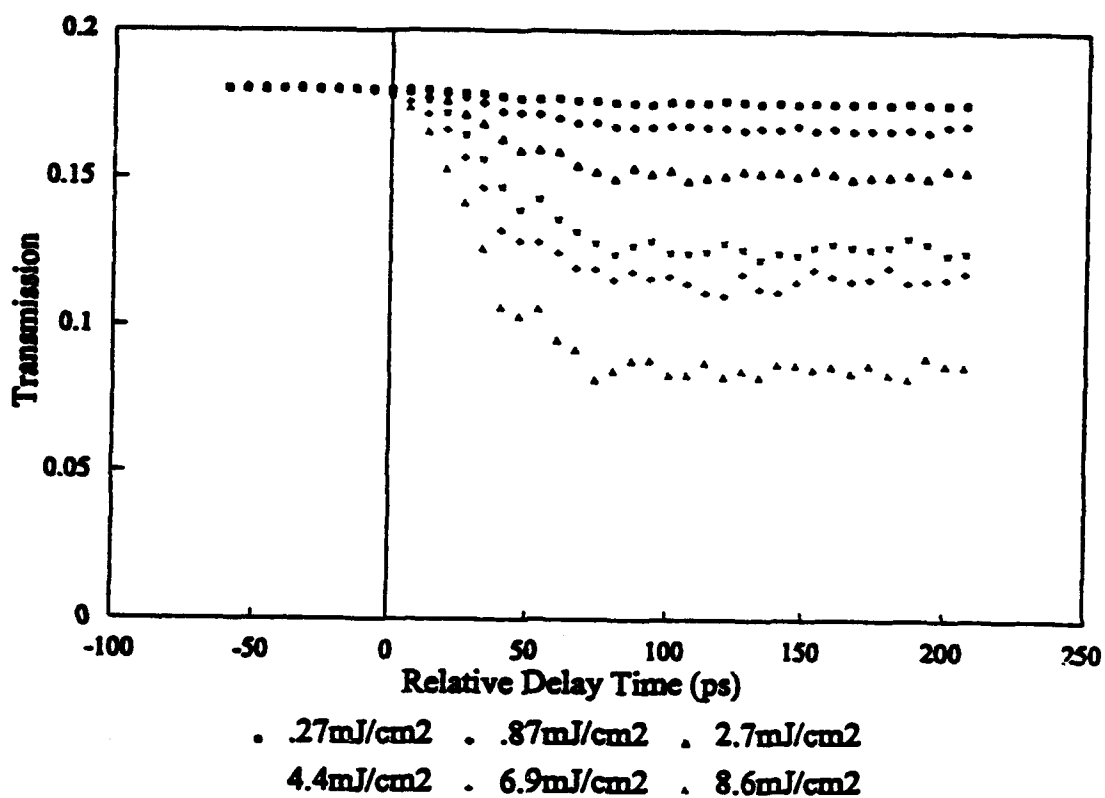


Fig. 2.3.1. Transmission coefficient of the probe beam through C<sub>60</sub>/toluene cell as a function of relative timing between the pump and probe pulses at various pump intensities.

ratio  $T_r$ , defined as [the delayed-probe-pulse transmission at low fluence + the delayed-probe-pulse transmission at fluence  $F$ ]. At fluence low enough to excite only a small fraction of  $C_{60}$  molecules, a simple model predicts that the logarithm of  $T_r$  is proportional to the fluence  $F$ . This relation is born out by our experimental results as shown here in Fig. 2.3.2. Unfortunately, the slope seen there predicts a reverse saturable absorption coefficient about several times stronger than that observed by Tutt and Kost (Tut92) and by Joshi, *et al.* (Jos93), using 532 nm pulses longer than 10 ns. Our results are in Talk 4.24 and are prepared for publication.

**2.3.4 Optical polymers.** We completed many new measurements of the nonlinear optical properties of polymers and pre-polymer molecules.. We continued studies to develop rugged and stable polymers for nonlinear optical imaging and real-time holography. This effort is in collaboration with professor L. R. Dalton in the USC Chemistry Department who synthesizes the materials and performs some initial analytical studies on them. We have also started collaboration with professor C. W. Spangler in the Department of Chemistry in Northern Illinois University. Our role in these projects is (1) to perform both linear and nonlinear optical measurements, (2) to identify the parameters and the processes to describe the nonlinearities, and (3) to take part in the identification of desirable physical properties for promising materials. During the current reporting period we have performed a number of experiments on a wide range of organic materials, as reported in Pubs. 3.2, 3.14, 3.15, 3.16, 3.17, and 3.26.

## **2.4 Photorefractive materials and properties**

We continued our contributions to photorefractive optics and materials, but at reduced efforts. We did achieve some long-standing goals as follows.

**2.4.1 Electron mobility in photorefractive  $Bi_{12}SiO_{20}$ .** During the current contract, we have developed what we believe to be the first method to measure directly the average drift velocity of photoexcited charge carriers in an insulator in a known electric field. We have also uncovered strong evidence that the photoexcited electrons spend large part of their migration time immobile,

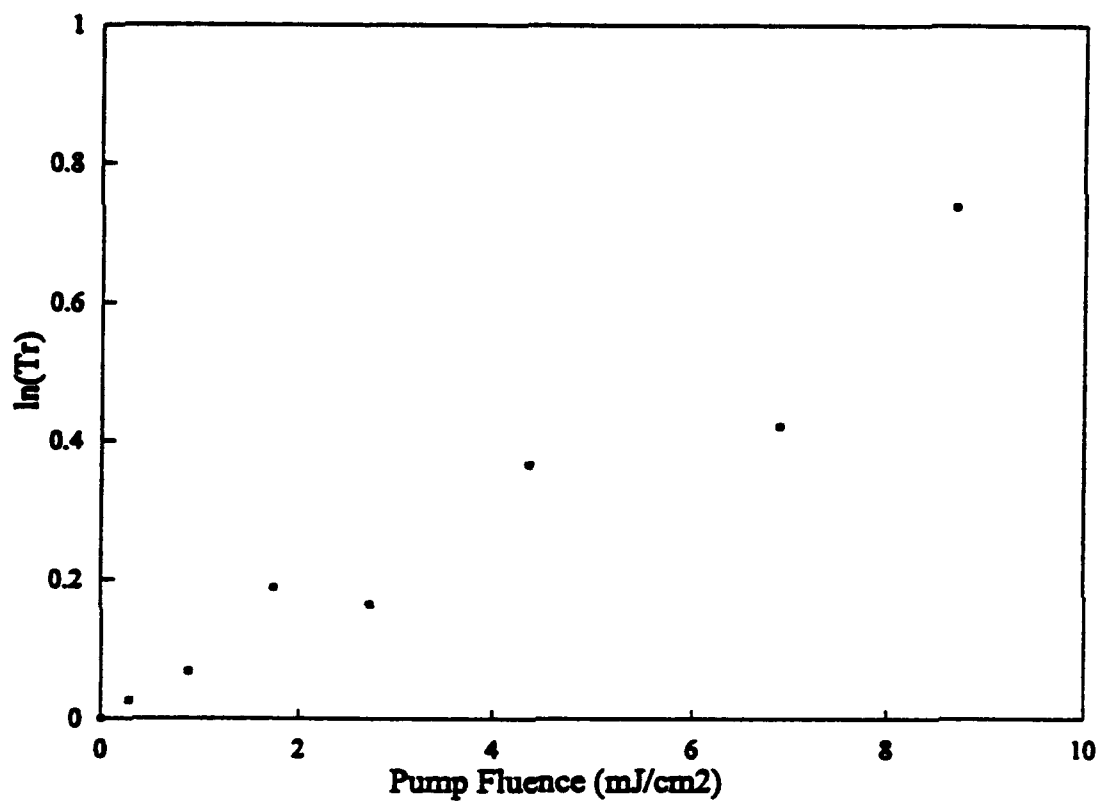


Fig. 2.3.2. Relative change in probe transmission through the C60/toluene cell caused by the pump pulse as a function of the incident pump fluence called F in the text.

dwelling for a large fraction their excited time span in shallow traps, before the final recombination with deep traps.

Details of our holographic time-of-flight measurements of mobility and comparison to other published measurements are given in Pubs. 3.1, 3.3, and 3.5. We found the electron drift mobility to be  $0.24 \pm 0.07 \text{ cm}^2/(\text{Vs})$  during the time of observation from 1  $\mu\text{s}$  to the recombination time of 80  $\mu\text{s}$ . We also determined the mobility as a function of temperature and observed an Arrhenius type temperature dependence with the activation energy  $320 \pm 40 \text{ MeV}$ , as we report in Pub. 3.10. The smallness of the mobility value and its Arrhenius type temperature dependence led us to develop and investigate a model in which the shallow traps limit the movement of photoexcited charge carriers. We found a simple analytical solution for this model in the limit of low excitation fluency which we believe describes our experiment. We feel more confident that we have found the correct physical description for transport of photoexcited electrons in  $\text{Bi}_{12}\text{SiO}_{20}$  because the analytical solution explains the previously unexplained damping of the oscillatory signal as a function applied electric field. This model and solution are reported as Pub. 3.18.

Our results suggested to us that we might be able to see the conduction band drift mobility directly within less than  $\sim 100 \text{ ns}$  after a picosecond excitation pulse. We have recently completed many such nanosecond-scale measurements of the photorefractive grating and verified that the conduction band mobility is an order-of-magnitude larger than the average mobility seen in  $\mu\text{s}$  experiments or slower. A typical direct measurement of the nanosecond-scale transient photorefractive effect is shown in Fig. 2.4.1 where one can read the drift velocity directly, as explained in the caption. These results appear in Thesis 5.3 and are in manuscript for journal submission.

**2.4.2 Optical polarizabilities of traps in photorefractive materials.** We have devised a novel grating technique to measure the optical polarizability difference between the occupied and vacant traps from which the carriers are photo-excited. We call this technique "transport-induced-grating interferometry." The imaginary part of the polarizability is directly related to the absorption cross section of the trap. The application of the method to (unidentified) traps in photorefractive  $\text{Bi}_{12}\text{TiO}_{20}$  was described in Talk 4.8 with a published summary. We have also determined the polarizability differences between occupied and

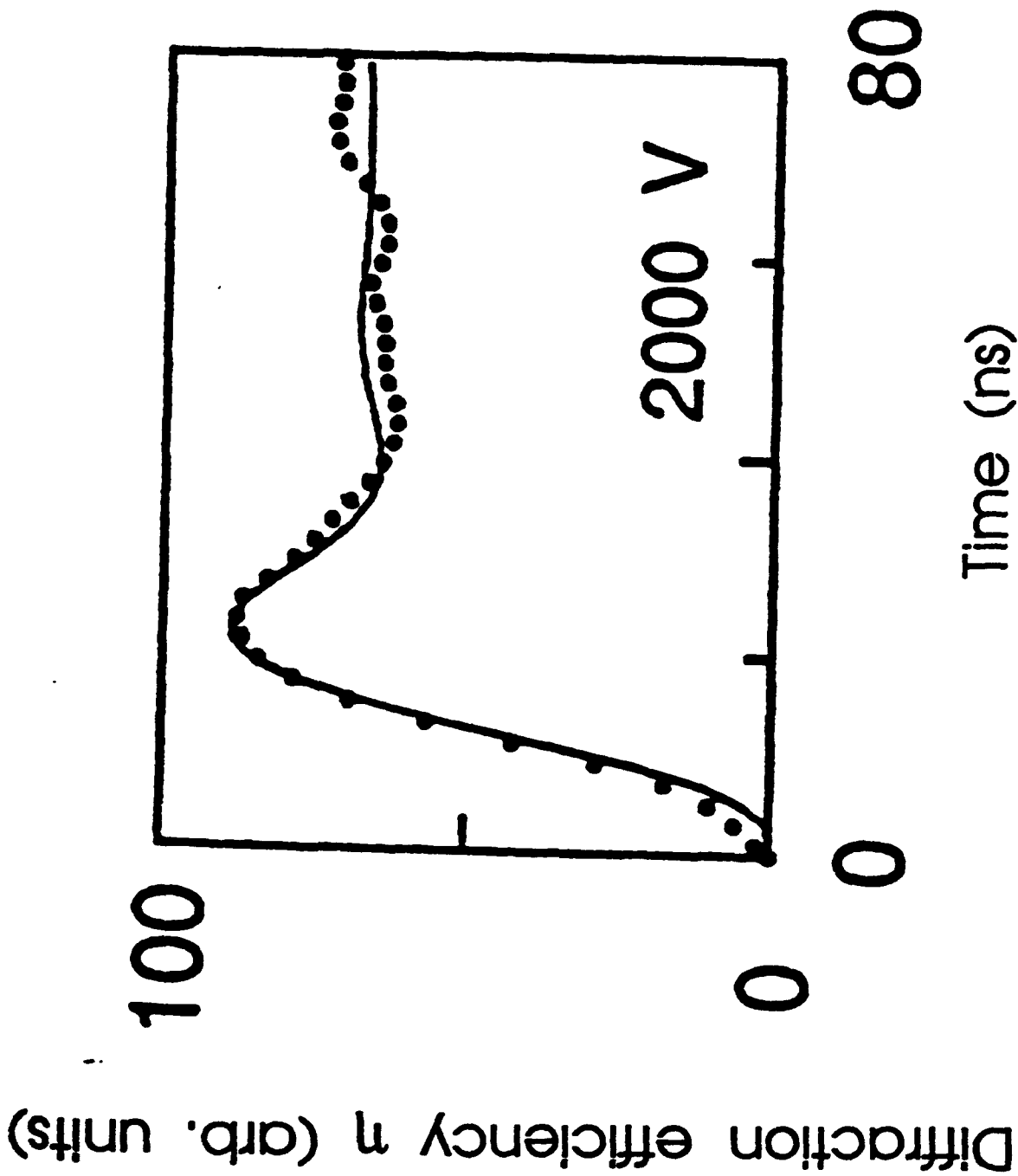


Fig. 2.4.1 Time-of-flight measurement of velocity of photoexcited electrons. Solid dots show our measured diffraction efficiency from the Coulomb charge grating formed by drifting photoelectrons excited by two  $80 \mu\text{J}/\text{cm}^2$ ,  $532 \text{ nm}$ ,  $30 \text{ psc}$  pulses intersecting at an angle of  $3.9$  degrees in cubic  $\text{Bi}_{12}\text{SiO}_{20}$ . Sample thickness was  $5 \text{ mm}$  so that an internal static uniform electric field of nearly  $4000 \text{ V}/\text{cm}$  was formed by the applied voltage of  $2000 \text{ V}$ . The delay of  $23 \text{ nsc}$  seen here before the first diffraction maximum is essentially the time required for the excited electrons to drift one half of the grating period ( $3.9 \text{ micron}$ ) determined by the beam intersection angle. This gives essentially the same electron drift mobility ( $\sim 4 \text{ cm}^2/\text{Vs}$ ) obtained by the best fit of our theory shown here by the solid line. The velocity was observed to be linear in the applied voltage up to breakdown fields ( $\sim 10 \text{ kV}/\text{cm}$ ).

vacant traps in photorefractive  $\text{Bi}_{12}\text{SiO}_{20}$  using a moving grating technique. These results are reported in Pub. 3.22.

**2.4.3 Spatial harmonics in photorefractive  $\text{BaTiO}_3$ .** We have made the first quantitative measurements of the spatial harmonic content of photorefractive gratings and compared these measurements with computer solutions of the standard model of photoexcitation, drift, diffusion, and direct recombination of a single carrier species. These experiments were carried out in  $\text{BaTiO}_3$ . We obtained a good fit between the experiments and the theory except in the case of strong fringe contrast ( $0.8 < m < 1$ ). These results are described in Pub. 3.6.

**2.4.4 Characterization of cw-laser-induced gratings.** We have invented a technique for complete characterization of cw-laser-induced volume gratings in dielectric materials. Laser-induced dielectric gratings are sinusoidal spatial variations in a material's dielectric function which are induced by the interference pattern of two laser beams and which can diffract light. They can have two components: a real refractive index grating and an absorption grating. In the general case of a noncentrosymmetric dielectric material, both the real refractive index grating and the absorption grating can have their maxima spatially shifted with respect to the optical interference patterns. Thus the full description of laser-induced dielectric grating requires four parameters: two amplitudes and two shifts. We have applied our method to find all four parameters in cw-laser-induced dielectric gratings in  $\text{KTaO}_3$  doped with Cu. We found a strong real refractive index grating ( $\Delta n \approx 2 \times 10^{-5}$ ) in phase with the intensity pattern as described in the published summaries of Talks 4.16 and 4.27.

## **2.5 Anomalous optical transmission in latex suspensions**

Having shown before the current contract period that highly scattering media can exhibit uncommonly large optical nonlinearity, we continued studies of such media. The major result of this period was our discovery of anomalous transmission in latex suspensions. We made what we believe is the first observation of the nonexponential attenuation of a weak optical beam in a homogenous medium, that is, the first exception to the famous property of weak light beams that was first enunciated by Pierre Bouguer more than 260 years ago: "In a medium of uniform transparency the light remaining in a collimated beam is an

exponential function of the length of its path in the medium." We expected such a violation would occur in our latex suspensions when the attenuation length due to scattering was of the order of (or shorter than) the correlation distance between the scattering particles. Our detailed theory fits the attenuation measurements well. Details of this research are given in Pubs. 3.4 and 3.7.

Our studies of nonexponential beam attenuation arose in the course of our long-standing project to study homogenous suspensions and mixtures of non-absorbing particles. These media exhibit large optical nonlinearity (Kerr effect) by virtue of physical processes (such as molecular reorientation) that also cause a large amount of light scattering. There is also the possibility that, if scattering is strong enough, randomly distributed localized light modes will form, similarly as "Anderson localization" of electronic states forms in random media. The nonexponential beam attenuation is related to light localization phenomena.

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### 3. PAPERS AND Ph.D. THESES PUBLISHED FROM THIS PROJECT

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- 3.3. J. P. Partanen, P. Nouchi, J. M. C. Jonathan, and R. W. Hellwarth, "Charge transport and holographic determination of the mobility of the charge carriers in photorefractive BSO," *Annales de Physique, Colloque no 1, Suppl. no 1*, vol. 16, pp. 135 - 142 (1991).
- 3.4. P. Tam, and R. W. Hellwarth, "Anomalous optical transmission in homogenous latex suspensions," *Phys Rev. B*, vol. 43, pp. 13314 - 13319 (1991).
- 3.5. J. P. Partanen, P. Nouchi, J. M. C. Jonathan, and R. W. Hellwarth, "Comparison between holographic and transient photo current measurements of electron mobility in photorefractive  $\text{Bi}_{12}\text{SiO}_{20}$ ," *Phys. Rev. B*, vol. 44, pp. 1487 - 1491 (1991).
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- 3.7. P. W. Tam, "Anomalous optical transmission in homogenous latex suspensions," Ph.D. Thesis, Department of Physics, University of Southern California, Los Angeles, CA (1991).
- 3.8. R. J. Knize and J. P. Partanen, "Comment on 'Large infrared nonlinear optical response of  $\text{C}_{60}$ '," *Phys. Rev. Lett.*, vol. 68, p. 2704 (1992).
- 3.9. P. Nouchi, "Studies of photo-excited and trapped electrons in cubic  $\text{Bi}_{12}\text{SiO}_{20}$ ," Ph.D. Thesis, Department of Electrical Engineering, University of Southern California, Los Angeles, CA (1992).
- 3.10. P. Nouchi, J. P. Partanen, and R. W. Hellwarth, "Temperature dependence of electron mobility in photorefractive  $\text{Bi}_{12}\text{SiO}_{20}$ ," *J. Opt. Soc. Am. B*, vol. 9, pp. 1428 -1431 (1992).
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- 3.17. C. S. Spangler, T. A. Kelleher, P.-K. Liu, L.-T. Cheng, N. Tang, R. W. Hellwarth, "Substituent effects in the design of new organic NLO materials," will appear in *Organic materials for Nonlinear Optics III*, R. A. Hann and D. Bloor, the Royal Society of Chemistry, Cambridge, U. K. (1993).
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- 3.26 C. W. Spangler, M. Q. He, J. Laquindanum, L. Dalton, N. Tang, J. Partanen, and R. Hellwarth, "Bipolaron formation and nonlinear optical properties in bis-thienyl polyenes," will appear in *Electrical, optical, and magnetic properties of organic solid state materials*, vol. 328 of the *Mat. Res. Soc. Symp. Proc. Ser.* (1993).

#### 4. TALKS, SEMINARS, WORKSHOPS AND CONFERENCES STEMMING FROM THIS PROJECT

- 4.1. "Atoms for logic," R. J. Knize, B. Ai, D. Glassner, R. W. Hellwarth, and J. P. Partanen, Paper K1530 at the annual Meeting of the Division of Atomic, Molecular, and Optical Physics of the American Institute of Physics, Washington, D. C., April 24, 1991, Abstract in *Bulletin of the American Physical Society*, vol. 36, p 1374 (1991).
- 4.2. "Temperature dependence of electron mobility in photorefractive  $\text{Bi}_{12}\text{SiO}_{20}$ ," P. Nouchi, J. P. Partanen, and R. W. Hellwarth, Paper CWD4 at the CLEO'91 Conference, Baltimore, Maryland, May 15, 1991, Abstract in *Digest of CLEO'91 conference*, Optical Society of America, Washington, DC, Technical digest series, vol. 10, pp. 234-235 (1991).
- 4.3. "Optical beam coupling by trap polarizability gratings in  $\text{Bi}_{12}\text{SiO}_{20}$ ," P. Xia, J. M. C. Jonathan, J. P. Partanen, and R. W. Hellwarth, Paper CWD6 at the CLEO'91 Conference, Baltimore, Maryland, May 15, 1991, Abstract in *Digest of CLEO'91 conference*, Optical Society of America, Washington, DC, Technical digest series, vol. 10, pp. 236 - 237 (1991).
- 4.4. "Spatial harmonics in photorefractive gratings, theory vs. experiment", R. W. Hellwarth, Clarendon Laboratory, Oxford, England, June 4, 1991.

- 4.5. "Atoms for logic," R. W. Hellwarth, Laboratoire de Physique des Lasers, Univ. of Paris-Nord, Villeteneuse, France, June 25, 1991.
- 4.6. "Atoms for logic," R. W. Hellwarth, Institut d'Optique, Orsay, France July 2, 1991.
- 4.7. "Thinking about laser gravity wave detectors," R. W. Hellwarth, 6th Annual Theoretical Physics and Quantum Optics Summer Festival, Max-Planck Institut fuer Quantenoptic (MPQ), Garching, Germany, July 11, 1991.
- 4.8. "Transport-induced-grating interferometry: application to photorefractive  $\text{Bi}_{12}\text{TiO}_{20}$ ," P. Xia, J. P. Partanen, and R. W. Hellwarth, Paper TuC8 at the Meeting on Photorefractive Materials, Effects, and Devices III, Beverly Massachusetts, July 30, 1991, Abstract in Technical digest of Meeting on Photorefractive Materials, Effects, and Devices III, Optical Society of America, Washington, DC, Technical digest series, vol. 14, pp. 228 - 231 (1991).
- 4.9. "Conduction band and trap limited mobilities in  $\text{Bi}_{12}\text{SiO}_{20}$ ," P. Nouchi, J. P. Partanen, and R. W. Hellwarth, Paper TuC8 at the Meeting on Photorefractive Materials, Effects, and Devices III, Beverly Massachusetts, July 30, 1991, Abstract in Technical digest of Meeting on Photorefractive Materials, Effects, and Devices III, Optical Society of America, Washington, DC, Technical digest series, vol. 14, pp. 236 - 239 (1991).
- 4.10. "Optical logic with single photons," R. W. Hellwarth, Bellcore Laboratories, Red Bank, New Jersey, October 22, 1991.
- 4.11. "Ground rotational states of  $^{12}\text{C}_{60}$  fullerene," J. P. Partanen, 22nd Winter Colloquium on Quantum Electronics, Snowbird, Utah, January 8, 1992.
- 4.12. "Maximum fieldstrength of pulsed solutions of Maxwell's equations in vacuum," P. Nouchi, 22nd Winter Colloquium on Quantum Electronics, Snowbird, Utah, January 8, 1992.
- 4.13. "Direct measurement of conduction band mobility in  $n$ -type  $\text{Bi}_{12}\text{SiO}_{20}$ ," P. Nouchi, J. P. Partanen, and R. W. Hellwarth, Paper CTuC7 at the CLEO'92 Conference, Anaheim, California, May 12, 1992, Abstract in Technical digest of CLEO'92 conference, Optical Society of America, Washington, DC, Technical digest series, vol. 12, pp. 84 - 85 (1992).
- 4.14. "Laser-assisted degenerate four-wave mixing in cesium," D. S. Glassner, J. M. C. Jonathan, B. Ai, R. J. Knize, R. W. Hellwarth, and J. P. Partanen, Paper QTuI1 at the QELS'92 Conference, Anaheim, California, May 12, 1992, Abstract in

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- 4.16. "Technique for characterization of laser-induced dielectric gratings: Application to non-photorefractive cubic  $\text{KTaO}_3\text{:Co}$ ," P. Xia, J. P. Partanen, and R. W. Hellwarth, Paper TuD26 at NLO'92 Conference, Lahaina, Maui, Hawaii, August 18, 1992, Abstract in Digest of NLO'92 conference, Optical Society of America, Washington, DC, Technical digest series, vol. 18, pp. 266-368 (1992).
- 4.17. "Large infrared nonlinear optical response of  $\text{C}_{60}$  not observed," N. Tang, J. P. Partanen, R. J. Knize, R. W. Hellwarth, M. McLean, and L. Dalton, Paper TuAA5 at OSA Annual Meeting, September 22, 1992, Abstract in Technical digest of OSA Annual meeting, Optical Society of America, Washington, DC, Technical digest series, vol. 23, p. 78 (1992).
- 4.18. "Can one build a tabletop TeV particle accelerator?" R. W. Hellwarth, Lawrence Livermore Laboratory, Livermore, California, Feb. 26, 1993.
- 4.19. "Energy and ability to accelerate charge of a new pulse solution of Maxwell's equations in vacuum", R. W. Hellwarth, Max. Planck Institut fur Quantenoptik, Garching, Germany, March 29, 1993.
- 4.20. "Acceleration of charged particles by an electromagnetic pulse in vacuum," R. W. Hellwarth, Physics & Astronomy Colloquium, University of Southern California, Los Angeles, April 26, 1993.
- 4.21. "Femtosecond response of optical Kerr materials: effects on degenerate four-wave-mixing," R. W. Hellwarth, Universite de Bordeaux I, Talence, France, July 8, 1993.
- 4.22. "Acceleration of charged particles by an electromagnetic pulse in vacuum," R. W. Hellwarth, Lab. de Physique des Lasers Seminar, U. Paris (Nord), France, July 21, 1993.
- 4.23. "Image Correlator Using Alkali Vapor," R. W. Hellwarth, Institut d'Optique, Orsay, France, July 27, 1993.
- 4.24. "Picosecond studies of the optical nonlinearity of  $\text{C}_{60}$  and  $\text{C}_{70}$ ," N. Tang, J. P. Partanen, and R. W. Hellwarth, 1993 Cordon Research Conference on Nonlinear

Optics and Lasers, Brewster Academy, Wolfeboro, New Hampshire, August 3, 1993

- 4.25. "Acceleration of charged particles by an electromagnetic pulse in vacuum: Can you build a TeV particle accelerator on a table top?" R. W. Hellwarth, Physics Departmental Colloquium, U. of Illinois, Urbana, Illinois, Sep. 9, 1993.
- 4.26. "Lasers in the Lab and in the Home," Freshman Colloquium, Physics Dept., USC, Sept. 28, 1993.
- 4.27. "Large cw-laser-induced grating in  $\text{KTaO}_3\text{:Co}$ ," P. Xia, J. P. Partanen, and R. W. Hellwarth, Paper MD1 at OSA Annual Meeting, October 4, 1993, Abstract in Technical digest of OSA Annual meeting, Optical Society of America, Washington, DC, 1993 Technical digest series, vol. 16, p. 5 (1993).
- 4.28. "Improved angular response of degenerate four wave mixing in K vapor," D. S. Glassner, B. Ai, R. J. Knize, J. P. Partanen, and R. W. Hellwarth, Paper MHH5 at OSA Annual Meeting, October 4, 1993, Abstract in Technical digest of OSA Annual meeting, Optical Society of America, Washington, DC, 1993 Technical digest series, vol. 16, pp. 38 - 39 (1993).
- 4.29. "Thin atomic Cs vapor as a nonlinear optical medium," B. Ai, D. S. Glassner, R. J. Knize, J. P. Partanen, and R. W. Hellwarth, Paper MHH6 at OSA Annual Meeting, October 4, 1993, Abstract in Technical digest of OSA Annual meeting, Optical Society of America, Washington, DC, 1993 Technical digest series, vol. 16, p. 39 (1993).

## **5. ADVANCED DEGREES AWARDED 12/1/90 - 11/30/93**

- 5.1. "Four-wave mixing in photorefractive and polymeric materials: ring phase conjugator and time-resolved measurement of third order optical nonlinearity," J. P. Jiang, Ph.D. Thesis (Physics), USC Dec. 1991.
- 5.2. "Anomalous optical transmission in homogenous latex suspensions", P. W. Tam, Ph.D. Thesis (Physics), USC, Dec. 1991.
- 5.3. "Studies of photo-excited and trapped electrons in cubic  $\text{Bi}_{12}\text{SiO}_{20}$ ," P. Nouchi, Ph.D. Thesis (Electrical Engineering), USC, May 1992.
- 5.4. "Study of laser-induced dielectric gratings in nonlinear optical crystals," Ping Xia, Ph.D. Thesis (Electrical Engineering), USC, May 1993.

## **6. PROFESSIONAL PERSONNEL 12/1/90 - 11/30/93**

- 6.1. Dr. R. W. Hellwarth
- 6.2. Dr. J. P. Partanen
- 6.3. Dr. J. M. C. Jonathan
- 6.4. Ms. Pascale Nouchi
- 6.5. Ms. Ping Xia
- 6.6. Mr. Patrick Tam
- 6.7. Mr. David Glassner
- 6.8. Mr. Nansheng Tang

## **7. INTERACTIONS 12/1/90 - 11/30/93**

The interactions by the members of this project are, in addition to the talks, conferences, workshops, and seminars listed in Section 4, the consulting services performed by Professor Hellwarth for Lawrence Livermore National Laboratory (contact: Dr. Mark Henesian), for Los Alamos National Laboratory (contact: Dr. D. F. Dubois), and for the Hughes Research Laboratory (contact: Dr. C. R. Giuliano). The subject matter for these consultations were lasers and laser beam interactions.

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